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Development of a Low-Level, In-Line Alpha Counter (LLILAC)

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Abstract

This is the final report of a two-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). With the increasing awareness of water contamination issues and the rising consequences of any form of contamination, real-time continuous monitoring is rapidly becoming a necessity. In particular, monitoring for the presence of any radioactive material is paramount. The most difficult of such monitoring tasks is that of detecting alpha-emitting radionuclides in water. Our development of the Low Level In-Line Alpha Counter (LLILAC) addresses the need for on-line, near real-time monitoring of alpha-emitting radionuclides in aqueous streams in a wide variety of applications. Although primarily designed as an on-line instrument for real-time applications, the detector can also be used for long-term *in situ*/post-closure monitoring. This detection system operates by allowing the stream to be monitored to come in contact with a large number of small rods or tubes made of scintillation material. By maximizing the surface to volume ratio of the scintillator, the response to alpha particles is favored over other types of radiation. Several configurations of scintillator and light collection schemes have been investigated to optimize the detection efficiency. We have also written several Monte Carlo codes to help to predict and understand the detector performance.

1. Background and Research Objectives

The United States is facing one of the largest cleanup efforts in history. The minimum estimated cost is 160 billion dollars over a thirty-year period, which anticipates the introduction of advanced technologies that are now under development in a timely fashion. The characterization activities associated with the cleanup currently cost the nation 1 billion dollars annually, with increases expected. Pressing monitoring issues include how to perform long-term post-closure monitoring in a cost-effective fashion. Of particular importance is the characterization and monitoring problem of detecting alpha and low-energy beta-emitting radionuclides in aqueous streams.

Many processes in the DOE complex as well as in the commercial sector involve aqueous streams containing radioactive materials. Examples of activities that produce such

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streams include fuel reprocessing, waste treatment facilities, and remediation activities. Increasing awareness of water contamination issues in the environment and rising consequences of any form of contamination is placing increased demands on effluent monitoring. In particular, monitoring for the presence of any radioactive material is paramount. However, one of the more challenging on-line instrumentation needs is for monitoring of alpha emitting radionuclides in aqueous process streams. Requirements to reduce analytical cost, and analytical response time make existing batch sampling techniques unfavorable when compared to on-line instrumentation that can yield near real-time analytical results.

Currently available technology requires that a sample of the aqueous stream -- be it waste, process solutions, or effluent -- be extracted and chemically processed for sample counting. Replacing conventional sampling and counting measurement techniques (such as liquid scintillation counting, whose scintillation cocktail poses waste disposal problems) with an on-line technique will eliminate the cost of batch-sample processing and chemistry for assay of alpha-producing radioactive materials. If verification of some process or remediation effort required thousands of samples -- at \$100 to \$200 per sample -- the savings could be several hundred thousand dollars for each process, with savings of millions over the life of the activity. An on-line technique also enhances safety and the as-low-as-reasonably-achievable principle, as no samples are removed or handled. Furthermore, conventional measurement techniques are far from real-time or on-line, as they may take days to weeks to complete. Of greater impact, however, is the increased public and regulatory confidence that process and effluent streams are being continuously monitored for escaping radioactivity. This increased public and regulator confidence will make some operations possible that were not previously.

The Low-Level, In-Line Alpha Counter (LLILAC) fulfills this need for on-line monitoring of alpha-emitting transuranic elements in aqueous streams. Adaptable for both process and effluent monitoring, the LLILAC is sensitive to alpha and beta emitting radionuclides. Although primarily designed as an on-line instrument for real-time applications, the detector can be used for long-term *in-situ*/post-closure monitoring of clear aqueous streams (for example, above- or below-ground natural water streams).

2. Importance to LANL's Science and Technology Base and National R&D Needs

This project supports a Los Alamos core competency in earth and environmental systems. It can help Los Alamos intensify the focus on specific environmental issues of concern to the public and also allow Los Alamos to address the environmental problems of the DOE Complex, DoD, and other governmental agencies. It enhances the Laboratory's visibility

in the area of environmental radioactivity monitors and increases LANL's ability to respond to initiatives in that area.

3. Scientific Approach and Results to Date

The LLILAC detector system uses a bundle of scintillation fibers or tubes fabricated such that the aqueous stream flows in the interstitial volume around the fibers or through the tubes. Light produced by the alpha particles striking the scintillation material is collected and processed to determine the concentration of alpha-emitting radionuclides present in the liquid. Initial estimates predict an on-line sensitivity of around 100 pCi/liter. This sensitivity is limited by the size of the detector and the detector background.

The detector background count rate is due primarily to cosmic-ray interactions. The 100 pCi/liter sensitivity estimate assumes no background suppression efforts. Anti-coincidence techniques are commonplace in low-level detection, and this detector's small size lends itself to active background suppression using such techniques. Since the detector consists of an array of thin scintillation fibers or tubes, some method of alternating the tube or fiber light collection would allow an additional background suppression technique. While cosmic-ray events will activate a number of fibers, an alpha event (at low rates) will only activate a single fiber. By using a coincidence veto, cosmic-ray events can be further discriminated against. Pulse shape and energy discrimination also provide possible avenues of background reduction.

The other significant technical challenge is the light collection design. The fibers must couple to a phototube in an efficient manner, while leaving enough open space to allow efficient fluid flow. The simplest approach arranges the fibers or tubes such that the aqueous solution couples light from the end of the fiber or tube directly to the photocathode of the photomultiplier tube.

Modeling of LLILAC Detector

As part of the evaluation of whether we can detect alpha particles in water we developed a model of the detector. Our basic modeling geometry consisted of either close-packed scintillation soda straws or close-packed scintillation rods. In the case of the straws, water could flow both inside and outside the straws. The solid rods or fibers are essentially straws where the flow is restricted to the outside. For the initial model we tracked alpha particles with a fixed initial energy and determined what percentage reached a scintillator when starting from a random position with a random direction vector within the water volume. We also tracked how much energy an alpha particle deposited in a scintillator and thus were able to construct energy histograms.

The primary design parameter we investigated was the radius of the straw, which has a strong effect on the overall efficiency. Since the range of alpha particles in water is only on the

order of 30 μm , only those particles generated in a thin layer near the surface of the scintillator will be detected. Based on this, a first approximation for the detection efficiency of alpha particles coming from a liquid contained inside a straw will be proportional to the ratio of the inside surface area to volume of the straw. This ratio is determined by the inside radius of the straw and is given by $2/r_i$, where r_i is the inside radius of the straw. Small diameter straws favor the detection of the alpha particles but also have higher resistance to flow of the aqueous media through the straws.

The ratio of interstitial volume to total bundle volume in close packed straws is independent of the straw diameter and is given by $1-(\pi/(4\sin 60^\circ))$. The interstitial area represents only 9.31% of the total bundle cross sectional area so that to achieve a given open interstitial area requires a large bundle of straws or rods. The advantage of the interstitial area is that the surface to volume ratio is larger than in the case for the interior of the straws. In this case, the ratio of the surface area to volume is given by $19.5/r_o$, where r_o is the outer radius of the straw or rod. When comparing the relative efficiencies of the interstitial volume with the internal volume of the straws, one concludes that for a given radius the interstitial efficiency will be some ten times larger than the internal efficiency (assuming very thin walls for the straws).

Other parameters we can alter include the thickness of the wall of the straw, its length, and the initial energy of the alpha particle. For flexibility the entire model was built as a set of objects in C++ so that we could alter geometries and media relatively quickly without having to alter the basic program.

Table 1 gives results for some of the detector configurations we modeled. Key observations are that overall efficiency does increase with smaller radius straws as discussed above and that for the diameters considered, the interstitial region between straws contributes a significant fraction of the detected activity even though it comprises a small fraction of the total volume. This result occurs because the geometry of the interstitial region is such that an alpha particle is never far from the outer wall of a straw. Unfortunately, use of this region is difficult because surface tension makes it difficult to flush water through these cavities when compared to flow through the interior of the straws.

Table 1: Three calculations for different inner and outer radii of straws. Reported are the fraction of water volume within a straw and associated efficiency, the fraction of water volume between straws and associated efficiency, and the total efficiency of the detector.

Case	Inner radius (cm)	Outer radius (cm)	Frac. vol inside straw	Efficiency	Frac. vol outside straw	Efficiency	Total Efficiency
1	0.05	0.13	0.75	0.039	0.24	0.12	0.059
2	0.08	0.15	0.83	0.027	0.17	0.10	0.040
3	0.10	0.18	0.86	0.017	0.14	0.09	0.028

The results did suggest that solid fibers rather than straws, could be investigated. Note however that even with higher efficiency, the total number of alpha particles detected is higher for the inside of the straw than in the interstitial region because the increased volume more than makes up for the decreased efficiency. Thus a solid fiber scintillator would be most appropriate for the case where we want the highest efficiency using the smallest amount of liquid.

Data Acquisition System

The Data Acquisition System (DAS) software provides control and monitoring of the LLILAC system. We have used a 486-based IBM-PC that is interfaced to a CAMAC crate via a DSP-6002 crate controller to collect counting data and control experiment operation. The LLILAC electronics system utilizes standard fast nuclear instrument modules for the processing of pulses from the photomultiplier tubes (PMTs). The data is then sent to CAMAC modules for data acquisition. High voltage for the PMTs is provided by a single LeCroy Model HV4032A high-voltage power system mainframe with a 4-channel, negative 3.5 kV plug-in module. The following block diagram illustrates the photomultiplier pulse processing electronics.

Based on previous experience, we expected rather low light intensities and have selected Burle-type 8850 PMTs for the light detection. These tubes have good timing performance and are capable of single photoelectron resolution. In our system, the small

current pulses from the PMTs are amplified by a factor of 200 using an ORTEC Model FTA-820 fast amplifier. The amplified pulses are then sent to constant-fraction timing discriminators and are converted to logic pulses. If both tubes register a light pulse within 50 nsec, then that is considered a real event and the charge signals from each tube are digitized and the event is counted. We decrease statistical uncertainty for each tube event by averaging the digitized data from the two tubes. The use of two tubes in coincidence mode is required at low light levels to eliminate thermally produced signals from the PMTs. In our typical operating conditions, each PMT generates thermally produced single-electron events at a rate of approximately 1 kHz. By requiring both tubes to generate a signal within 50 nsec for a real event, we reduce the random thermally produced counting rate from a total of 2 kHz for both tubes with no coincidence requirements to approximately 0.1 Hz. The cosmic-ray generated background is approximately 15 Hz, so the random thermally generated rate represents less than 1% of the background rate.

The executable code, LLILAC, was compiled with a National Instruments Lab Windows/CVI compiler and is a Microsoft Windows-based executable. The LLILAC program allows the user to access the DAS system with functional interface panels within the Microsoft Windows environment. The areas of control and monitoring available are system initialization, tube spectral analysis, and high voltage monitoring and control.

When LLILAC is launched, a dialog box allows the user to select a default configuration file or the name of a different configuration file to load. The PMT configuration file contains all the parameters associated with the operation of the photomultiplier tubes and the electronics system configuration. The file can be modified for a new setup with any ASCII text editor. During the initialization phase, the LLILAC program will read the slot location and total number of CAMAC modules in the system. Photomultiplier tubes are identified by name, channel, and pair locations in the system. After the tubes have been identified, the software will set the location of each tube ADC and high-voltage channels for each PMT.

The tube-spectrum panel displays the histogram of the charge data collected from each of the 8850 photomultiplier tubes in the system. The display of the charge spectrum from each tube can be selected individually after a collection run has been completed. The data collection period is specified by the number of tube events that have occurred above the hardware discriminator threshold on the EG&G CF8000 and the user specified "events/run." The discriminator threshold is usually set to channel 20 (approximately 5 pC) in the charge spectrum plot to eliminate low level noise. The high voltage for the PMTs is adjusted so that the single photoelectron signals generate approximately 11 pC of charge and are stored in channel 45 in the charge spectra. The single-photoelectron events provide an absolute system calibration for the number of photoelectrons produced in a scintillation count. The data acquisition system will continue to collect tube events and plot the histogram every 500 events until the user-specified "events/run" has been obtained. At the completion of an event run, the user has the option of rescaling the plot in either of the axes and making a choice of linear or

logarithmic plots. Two green-line plot cursors are used to select a region of interest for integration measurements and, during a run, a red cross-hair cursor tracks the histogram peak. Tube-spectrum data can be saved in a user-specified file for post processing and analysis. The file stores the date, time, user comments, and the tube histogram data in a comma-delimited Excel file format.

Experimental Test Description

In all of the detector configurations to date we have used scintillation tubes supplied by Bicron and made from BCF-10 scintillation plastic. The tubes are nominally 3 mm o.d. and 1 mm i.d.. We had hoped to obtain tubes with much thinner walls, but we were not able to find commercial product other than the Bicron-supplied material. Clearly, thinner-walled material would decrease the background and improve the beta particle rejection.

A typical test geometry is shown in Figure 1. In the case illustrated, the scintillation tubes are contained in an acrylic tube and the two acrylic light guides serve to collect the light from the sides of the straw array. In an effort to improve light collection we have also made a version of the detector in which the acrylic light guides contain the scintillation tubes directly.

Figure 1. Typical implementation of the Low Level In-Line Alpha Counter (LLILAC).

A 1-cm diameter hole in the light-guide halves provides a port into the interstitial volume and provides access for alpha- or beta-source testing of the detector. In the tests made to date, we have not introduced aqueous radioactive material in the interstitial volume because

of concerns about the difficulty in removing all of the activity after the tests were completed. For flow tests through the tubes, we used a flow rate of 1.5 l/min.

The scintillator tube array for a slightly different test cell is shown in Figure 2. In this cell, the straws are contained within a 12.75 in. x 2 in. o.d. clear acrylic tube and are supported inside the casing with three aluminum support disks. Each disk supports sixty evenly distributed straws set on four radii. Pipe fittings installed perpendicular to the casing at each end are for the water inlet and outlet ports. A silicone rubber plug cast at each end of the detector cell forces water flow through the scintillator tubes and prevents flow through the interstitial volume. Photomultiplier tubes are coupled to the windows at each end of the detector with RTV-615 silicone rubber. We have used this cell to evaluate the effect that water in the interstitial region has on the light collection from the ends of the scintillator array and to compare light collection from the ends of the array with light collection from the sides of the array.

Figure 2. End view of alternate spacing scheme for LLILAC scintillating tubes.

Discussion of Experimental Tests

The response of the current version of the detector is shown in Figure 3, where we show typical energy spectra for a high-energy beta-emitting nuclide ($^{90}\text{Sr}/^{90}\text{Y}$), for an alpha-emitting nuclide (^{241}Am), and the detector background.

These data were collected using light guides that collected the light from the sides of the scintillation tube array and the radioactive sources were external to the tube array. Counting was done in such a way that there is the same number of counts in each spectrum so that the

statistical uncertainty in each spectrum is similar. In the case for ^{90}Sr , a typical smooth beta spectrum is produced except that there is a low-energy peak corresponding to single photoelectron production. For ^{241}Am , there is a fairly broad peak due to the 5.4 MeV alpha emission and again there is the low energy peak associated with single photoelectron detection. From these measurements, we can estimate the relative light production from alpha and beta radiation. For the ^{90}Y daughter of ^{90}Sr with a maximum energy of 2.2 MeV, the response extends out to approximately channel 500 corresponding to approximately 11 photoelectrons (pe) or approximately 5 pe/MeV. For the alpha particles, the peak is at channel 150 corresponding to approximately 3.3 pe or approximately 0.6 pe/MeV. This relative light production is typical of plastic scintillators and would indicate that the detector should be better at detection of beta particles than alpha particles. We can compensate for this by using very thin scintillators so that, while the alpha particles will lose all of their energy in the detector, beta particles will leave only a small fraction of their energy in the detector.

We have also measured the alpha detection efficiency for aqueous radioactive solutions by filling one of the tubes with a solution of ^{241}Am . We sealed this tube and then could position the tube in the array. An alpha-particle detection efficiency of about 3% was measured using this tube. This measured value is in good agreement with our Monte Carlo estimates.

When we collected light from the sides of the array, we found that when we filled the interstitial volume with water to provide optical coupling, we collected about twice as much light as when the volume was left empty. It seems likely the light collection could be improved more if the interstitial volume was filled with a liquid that matched the index of refraction of the plastic more closely than water.

Our current detector has an active volume of only about 30 cm^3 and a total spectrum background of approximately 18 counts/sec. The background is dominated by single photoelectron events but there are some events recorded that have energies corresponding to 3-4 MeV equivalent beta particle energies.

We can estimate the detection limit by calculating the concentration of alpha emitting activity in the active volume of the counter necessary to have an alpha count rate equal to the background. For the current detector, the concentration would be approximately 580 nCi/liter. This concentration can be significantly reduced by building a larger detector and by keeping only those counts with energies corresponding to alpha events.

This approach to a novel alpha monitor has been shown to be workable and has an adequate detection sensitivity for a number of applications, especially in process control and monitoring. The mechanical design is quite flexible allowing different configurations to meet a large number of application-specific requirements.

Figure 3. Response of the LLILAC Detector to ^{90}Sr , ^{241}Am , and background.